Logarithmic susceptibility and optimal control of large fluctuations

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We analyze the probabilities of large infrequent fluctuations in systems driven by external fields. In a broad range of the field magnitudes, the logarithm of the fluctuation probability is linear in the field magnitude, and the response can be characterized by a *logarithmic susceptibility*. This susceptibility is used to analyze optimal control of large fluctuations. For nonadiabatic driving, the activation energies for nucleation and for escape of a Brownian particle display singular behavior as a function of the field shape.

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It was pointed out by Debye [1] that systems with coexisting metastable states may strongly respond to the driving field through the effect of the field on the probabilities of transitions between the states. For classical systems, the transition probability W is described by the activation law, $W \propto \exp(-R/kT)$. Even a relatively weak ac field h, for which the change of the activation energy $\Delta R \propto h$ is much less than R, can strongly affect W provided $|\Delta R|/kT$ is not small. This effect has been investigated for various systems and has attracted much attention recently in the context of stochastic resonance [2]. For $|\Delta R|/kT \gg 1$ the modulation of W becomes exponentially strong. So far this modulation has been analyzed for adiabatically slow driving, where the change of the field over the relaxation time of the system is small and fluctuational transitions occur "instantaneously", for a given value of the field (cf. [3]). The physical picture of transitions is different for *nonadiabatic driving*. In this Letter we provide nonadiabatic theory of large fluctuations in spatially extended and lumped parameter systems. We show that the exponentially strong dependence of the fluctuation probabilities on the driving field can be described in terms of an *observable* characteristic, the *logarithmic susceptibility* (LS).

The notion of LS and the way to evaluate it are based on the idea of the optimal fluctuational path. This is the path along which the system moves, with overwhelming probability, when it fluctuates to a given state or escapes from a metastable state. The distribution of fluctuational paths to a given state peaks sharply at the optimal path, as first noticed in [4].

Optimal paths in lumped parameter dynamical systems driven by Gaussian noise have attracted much theoretical interest [5] and were recently observed in experiments [6]. The notion of an optimal path applies also to continuous systems. Time-dependent fluctuations of the order parameter $\eta(\mathbf{x})$ were discussed in [7], and its optimal paths were considered in [8,9]. Optimal paths are "fluctuational counterparts" of dynamical trajectories: they map the problem of large noise-induced fluctuations onto the problem of noise-free dynamics of an auxiliary system (this dynamics depends on the properties of the noise driving the original system, see [5]).

We will first consider optimal paths and logarithmic susceptibility using as an example systems with a nonconserved order parameter [7](a). In these systems, fluctuations are described by the Langevin equation

$$\frac{\partial \eta(\mathbf{x},t)}{\partial t} = -\frac{\delta F}{\delta \eta(\mathbf{x},t)} + \xi(\mathbf{x},t), \qquad (1)$$
$$\langle \xi(\mathbf{x},t)\xi(\mathbf{x}',t')\rangle = 2kT\delta(\mathbf{x}-\mathbf{x}')\delta(t-t'),$$

with the free energy

$$F[\eta] = \int d\mathbf{x}\, \left[\frac{1}{2}\left(\boldsymbol{\nabla}\eta\right)^2 + V(\eta) - h(\mathbf{x},t)\eta\right], \qquad (2)$$

where $V(\eta)$ is the biased Landau potential.

The probability density for the system to fluctuate to a state $\eta_f \equiv \eta_f(\mathbf{x})$ at a time t_f is described by the activation law, $W[\eta_f; t_f] \propto \exp(-R[\eta_f; t_f]/kT)$, with the activation energy given by the solution of the variational problem [5,8]

$$R[\eta_f; t_f] = \min \frac{1}{4}\int_{-\infty}^{t_f} dt' \int d\mathbf{x}' \left[\frac{\partial \eta}{\partial t'} + \frac{\delta F}{\delta \eta}\right]^2. \qquad (3)$$

Here, the minimum is taken with respect to the paths $\eta(\mathbf{x}',t')$ that start from the stable state $\eta_{\rm st}(\mathbf{x},t)$ at $t' \to -\infty$, and arrive at the final state $\eta_f(\mathbf{x})$ for $t' = t_f$. We consider pulsed or periodic driving fields h, in which cases the state $\eta_{\rm st}$ is stationary (it provides a minimum to $V(\eta)$) or periodic, respectively.

Eq. (3) defines the action for an auxiliary Hamiltonian system, with the Lagrangian given by the integrand in (3). Extreme paths of this system that minimize R are optimal fluctuational paths $\eta(\mathbf{x},t)$ of the original system (1).

In the absence of driving the system (1) is in thermal equilibrium, and the activation energy is $R \equiv R^{(0)} = F^{(0)}[\eta_f] - F^{(0)}[\eta_{\rm st}^{(0)}]$ (the superscript 0 refers to the case $h=0$). The optimal fluctuational path $\eta^{(0)}(\mathbf{x},t|\eta_f,t_f)$ to the state η_f is the time-reversed path of (1) from this state in the neglect of noise, $\dot\eta^{(0)} = \delta F^{(0)}/\delta\eta$. This symmetry with respect to time reversal is a generic feature of systems with detailed balance [10].

To the first order in h, the field-induced change of R is given by the term $\propto h$ in the integrand in (3) evaluated along the unperturbed optimal path $\eta^{(0)}$:

$$R^{(1)}[\eta_f;t_f] = \int_{-\infty}^{t_f} dt' \int d\mathbf{x}' \chi(\mathbf{x}', t_f - t'|\eta_f) h(\mathbf{x}', t'), \quad (4)$$

$$\chi(\mathbf{x}, -t|\eta_f) = -\dot\eta^{(0)}(\mathbf{x}, t|\eta_f, 0) \quad (t<0). \quad (5)$$

The quantity χ describes the change $\propto h$ of the *logarithm* of the probability density to reach the state $\eta_f \equiv \eta_f(\mathbf{x})$, $\Delta \ln W \approx -R^{(1)}/kT$. This change may be *large*, and χ may be reasonably called the logarithmic susceptibility (LS). Like standard generalized susceptibility, LS has a causal form: the probability to reach a given state at a time t_f is affected by the values of the field at $t < t_f$. The LS $\chi(\mathbf{x}, t_f - t|\eta_f)$ becomes small for $t_f - t$ larger than the relaxation time of the system $t_{\rm rel}$. We note that Eq. (4) suggests how to *measure* LS for various states $\eta_f(\mathbf{x})$.

Of special interest are effects of the field on the probability of escape from a metastable state of the system. For a system (1) escape occurs via nucleation. For $h=0$, the critical nucleus $\eta_{\rm cr}^{(0)}(\mathbf{x}-\mathbf{x}_c)$ is the unstable stationary solution of the equation $\dot\eta = -\delta F^{(0)}/\delta\eta$ (the saddle point in the functional space), and the position of the center of the nucleus $\mathbf{x}_c$ is arbitrary. In forming the critical nucleus the system is most likely to move along the optimal path $\eta_{\rm nucl}^{(0)}(\mathbf{x}-\mathbf{x}_c, t-t_c) = \eta^{(0)}(\mathbf{x}-\mathbf{x}_c, t-t_c|\eta_{\rm cr}^{(0)}, \infty)$.

The optimal nucleation path $\eta_{\rm nucl}^{(0)}$ is a real-time instanton solution of the variational problem (3) for $h=0$. It starts for $t\to-\infty$ at the stable state $\eta_{\rm st}^{(0)}$ and approaches the state $\eta_{\rm cr}^{(0)}$ as $t\to\infty$. It is important that, as in the case of "conventional" instantons [11], $|\dot\eta_{\rm nucl}^{(0)}(\mathbf{x}-\mathbf{x}_c, t-t_c)|$ is large only within a time interval $|t-t_c| \lesssim t_{\rm rel}$ centered at an arbitrary instant t_c.

Translational and time degeneracy of the optimal nucleation paths $\eta_{\rm nucl}^{(0)}(\mathbf{x}-\mathbf{x}_c, t-t_c)$ is lifted when the system is driven by an external field. One can show that the optimal path still starts from the vicinity of the metastable state $\eta_{\rm st}$ and asymptotically approaches the critical nucleus $\eta_{\rm cr}$. In the case of a pulsed field, these states are just $\eta_{\rm st}^{(0)}$ and $\eta_{\rm cr}^{(0)}$, respectively. For weak periodic field, both states are periodic with the period of the field.

Lifting of the degeneracy occurs because the critical nucleus is an *unstable* state. For small h and finite $|t-t_c|$, the field-induced correction to a solution $\eta_{\rm nucl}^{(0)}(\mathbf{x}-\mathbf{x}_c, t-t_c)$, with given $\mathbf{x}_c, t_c$, remains small. However, generally the correction has an admixture of the solutions of the unperturbed Euler-Lagrange equations which go away from the unstable state exponentially in time. Therefore the whole solution diverges from the unstable state exponentially as $t\to\infty$. Only for one solution (or one per period of the field, in the case of periodic field) the divergence does not occur, and this solution is close to $\eta_{\rm nucl}^{(0)}(\mathbf{x}-\mathbf{x}_c, t_f-t_c)$ with certain $\mathbf{x}_c, t_c$ which are determined by the field. This situation is familiar from the Mel'nikov's theory of a heteroclinic orbit in the presence of a periodic perturbation [12].

An insight into the problem can be obtained from the analysis of the Lagrangian manifold of the Hamiltonian system with the action $R[\eta;t]$ (3). This manifold is formed by extremal trajectories with the generalized coordinate $\eta(\mathbf{x},t)$ and momentum $\pi(\mathbf{x},t) \equiv \delta R[\eta;t]/\delta\eta$ [12]. The optimal nucleation trajectory emanates, for $t\to-\infty$, from the stationary (or periodic, for periodic driving) state $(\eta_{\rm st}, \pi_{\rm st})$, which corresponds to the stable state of the original system (1), $\pi_{\rm st}(\mathbf{x},t)=0$. For $t\to\infty$ this trajectory approaches the state $\eta_{\rm cr}(\mathbf{x},t), \pi_{\rm cr}(\mathbf{x},t)=0$, associated with the critical nucleus.

Taking account of the first-order correction to the action (4), the equations for the first-order correction to an unperturbed extreme trajectory $\eta^{(0)}(\mathbf{x},t)$, $\pi^{(0)}(\mathbf{x},t) \equiv \dot\eta^{(0)}(\mathbf{x},t)$ take the form

$$\dot\eta^{(1)} = \mathbf{M}^{(0)}\eta^{(1)} + h + 2\frac{\delta R^{(1)}}{\delta\eta}, \quad (6)$$

$$\pi^{(1)} = \mathbf{M}^{(0)}\eta^{(1)} + \frac{\delta R^{(1)}}{\delta\eta},\ R^{(1)} \equiv R^{(1)}[\eta^{(0)}(\mathbf{x},t);t],$$

$$\mathbf{M}^{(0)}\eta^{(1)}(\mathbf{x},t) \equiv \int d\mathbf{x}' \frac{\delta^2 F^{(0)}}{\delta\eta(\mathbf{x})\delta\eta(\mathbf{x}')}\eta^{(1)}(\mathbf{x}',t).$$

The nucleation trajectory is close to an unperturbed path $\eta_{\rm nucl}^{(0)}(\mathbf{x}-\mathbf{x}_c, t-t_c)$ with properly chosen $\mathbf{x}_c, t_c$. The values of $\mathbf{x}_c, t_c$ can be found by solving Eqs. (6) near the critical nucleus, i.e. for large t, taking into account that, for the "correct" trajectory, the momentum $\pi^{(0)}+\pi^{(1)} \to 0$ for $t\to\infty$, while $\eta^{(1)} \to \eta_{\rm cr} - \eta_{\rm cr}^{(0)} \sim h$.

The solution of (6) near the nucleus can be expressed in terms of the eigenvalues λ_n and eigenfunctions $\psi_n(\mathbf{x}-\mathbf{x}_c)$ of the Hermitian operator $\mathbf{M}_{\rm cr}^{(0)} \equiv \mathbf{M}^{(0)}[\eta_{\rm cr}^{(0)}]$. The corresponding eigenvalue problem coincides with that for the dynamical equation (1) in the absence of noise, but the eigenvalues have the opposite signs. The operator $\mathbf{M}_{\rm cr}^{(0)}$ has one nondegenerate negative eigenvalue $\lambda_0 < 0$, with the eigenfunction

$$\psi_0(\mathbf{x}-\mathbf{x}_c) = C_0 e^{-\lambda_0(t-t_c)}\dot\eta_{\rm nucl}^{(0)}(\mathbf{x}-\mathbf{x}_c, t-t_c) \quad (7)$$

(here, $t\to\infty$); a degenerate zero eigenvalue $\lambda_1 = 0$, with the eigenfunctions which are proportional to the components of the vector $\boldsymbol{\nabla}\eta_{\rm cr}^{(0)}$; and positive eigenvalues $\lambda_n > 0$, $n>1$ [11,13].

It follows from Eqs. (4), (7) that, for $R^{(1)}$ evaluated along the path $\eta^{(0)}_{\rm nucl}$, the matrix element of $\delta R^{(1)}/\delta\eta$ on ψ_0 is equal to $A(\mathbf{x}_c,t_c)\exp[-\lambda_0(t-t_c)]$. It diverges for $t\to\infty$ unless $A=0$. It follows also from (6) that, in order for $\pi^{(1)}$ to go to zero for $t\to\infty$, the matrix elements of $\delta R^{(1)}/\delta\eta$ on $\partial\eta^{(0)}_{\rm cr}/\partial x_i$ should be equal to zero as well. The condition that the matrix elements should vanish is a consequence of the unperturbed system being "soft" in the functional-space directions ψ_0 and $\partial\eta^{(0)}_{\rm cr}/\partial x_i$ which correspond to the shifts of the nucleation path along t and $\mathbf{x}$. Taking account of Eqs. (4), (7), this condition can be written in the form

$$\frac{\partial\tilde R^{(1)}_{\rm nucl}(\mathbf{x}_c,t_c)}{\partial\mathbf{x_c}}=0,\quad \frac{\partial\tilde R^{(1)}_{\rm nucl}(\mathbf{x}_c,t_c)}{\partial t_c}=0, \tag{8}$$

$$\tilde R^{(1)}_{\rm nucl}\equiv\int_{-\infty}^{\infty}dt\int d\mathbf{x}\,\chi_{\rm nucl}(\mathbf{x}-\mathbf{x}_c,t-t_c)h(\mathbf{x},t)$$

$$\chi_{\rm nucl}(\mathbf{x}-\mathbf{x}_c,t-t_c)=-\dot\eta^{(0)}_{\rm nucl}(\mathbf{x}-\mathbf{x}_c,t-t_c) \tag{9}$$

Eqs. (8) determine $\mathbf{x}_c,t_c$ for the optimal nucleation path. They may have several roots. The physically relevant root is the one that provides the minimum to the field-induced correction to the activation energy of nucleation $R^{(1)}_{\rm nucl}$. According to Eqs. (4), (8),

$$R^{(1)}_{\rm nucl}=\min_{\mathbf{x}_c,t_c}\tilde R^{(1)}_{\rm nucl}(\mathbf{x}_c,t_c) \tag{10}$$

Eqs. (8) - (10) provide the nonadiabatic theory of nucleation rate. They have a simple physical meaning: in the presence of a time- and coordinate-dependent field, the optimal fluctuation finds the "best" time t_c and place $\mathbf{x}_c$ to occur. For thermal equilibrium systems, the correction is given by the work done by the field along the optimal path.

The correction $R^{(1)}_{\rm nucl}$ is linear in the magnitude of the driving field. However, the superposition principle does not apply: the correction from a sum of the fields is not equal to the sum of the corrections. This is because the minimum in Eq. (10) is a nonlinear operation, and the instant t_c and the position $\mathbf{x}_c$ of the optimal fluctuation are found in such a way as to minimize $R^{(1)}_{\rm nucl}$ for the total field, not for its constituents, and in particular not for each of its Fourier components.

We note that the dependence of $R^{(1)}_{\rm nucl}$ on the *shape* of the field may be singular. With the varying interrelation between the Fourier components of the field there occurs *switching* between different coexisting solutions of Eqs. (8), i.e. from one minimum of $\tilde R^{(1)}_{\rm nucl}$ to another, with different $\mathbf{x}_c,t_c$ (cf. inset to Fig. 1).

For pulsed fields, $R^{(1)}_{\rm nucl}$ is always non-positive: if the pulse effectively lowers (dynamically) the nucleation barrier (i.e., $R^{(1)}_{\rm nucl}<0$), the optimal fluctuation occurs where and when the field is "on", otherwise it follows from Eqs. (8), (10) that nucleation is most likely to occur where there is no field. The term $R^{(1)}_{\rm nucl}$ can be positive only provided the field has an appropriate time-independent (dc) component.

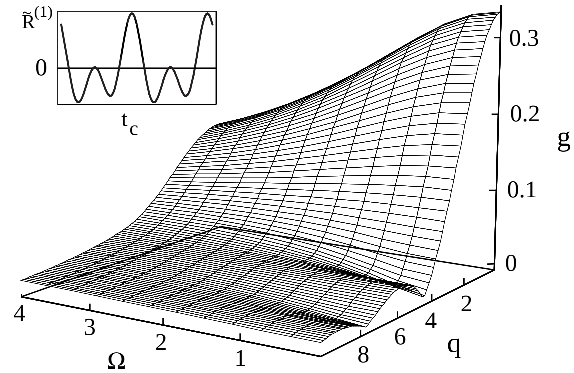

FIG. 1. Reduced absolute value of the logarithmic susceptibility for nucleation $g(\mathbf{k},\Omega)$ (11) in the case of a weakly asymmetric potential V. Inset: the correction $R^{(1)}(t_c)$ for nucleation in a uniform field $h=\cos\rho_c^2t+1.3\cos(2\rho_c^2t-0.1)$.

Analytical results for the logarithmic susceptibility for nucleation $\chi_{\rm nucl}$ (9) can be obtained in limiting cases, in particular for a weakly asymmetric double-well potential $V(\eta)=\frac14u\eta^4-\frac12r\eta^2-H\eta$, $|H|\ll r^{3/2}u^{-1/2}$. The critical nucleus in this case is a thin-wall droplet of a nucleating phase [11,14]. The optimal nucleation path corresponds to the increase of the radius of the droplet ρ until it reaches the critical value ρ_c, and is described by the time-reversed collapse [14] of the droplet in the absence of fluctutions. The resulting expression for the Fourier transform $\tilde\chi_{\rm nucl}(\mathbf{k},\omega)$ of the logarithmic susceptibility $\chi_{\rm nucl}(\mathbf{x},t)$ is of the form:

$$\tilde\chi_{\rm nucl}(\mathbf{k},\omega)\equiv\frac{6R^{(0)}_{\rm nucl}}{|H|}g\left(\rho_ck,\rho_c^2\omega\right), \tag{11}$$

$$g(q,\Omega)=\int_0^1dz\,\frac{\sin q\,z}{q}e^{-i\Omega z/2}(1-z)^{-i\Omega/2}.$$

(the free energy of the critical droplet $R^{(0)}_{\rm nucl}$ and ρ_c are given in [11,14]).

One can see from (9), (10) that, for a field of the form of a running or standing sinusoidal wave, $h=h_0\cos(\mathbf{kx}-\omega t)$ or $h=h_0\cos\mathbf{kx}\cos\omega t$, the correction to the activation energy is $R^{(1)}=-|\tilde\chi_{\rm nucl}(\mathbf{k},\omega)|h_0$. The susceptibility $|\tilde\chi_{\rm nucl}|$ as given by (11) is shown in Fig. 1.

The notion of logarithmic susceptibility makes it possible to formulate, in fairly general terms, the problem of *optimal control* of large fluctuations by an external field, or equivalently, of *cooperation* with fluctuations in bringing the system to a given state by a field which is much weaker than the one that would be necessary in the absence of fluctuations. In optimal control,

one has to minimize or maximize the activation energy $R[\eta_f; t_f] = R[\eta_f; t_f|h]$ (3) of reaching a state η_f in the presence of the field h subject to a given constraint on the field, i.e. to a given value of the *penalty functional* $G[h] = \mathcal{G}$ (for example, the energy in the field pulse).

The optimal activation energy $R_{\rm opt}$ and the corresponding optimal control field $h_{\rm opt}(t)$ can be found from the variational problem for the field

$$\delta \left[R[\eta_f; t_f|h] + \lambda \left(G[h] - \mathcal{G}\right)\right] = 0. \qquad (12)$$
$$R_{\rm opt} = R[\eta_f; t_f|h_{\rm opt}]$$

where λ is the Lagrange multiplier.

For various optimal control problems in physics and chemistry the penalty functional G is quadratic in h [15]. Then, for comparatively weak fields where the field-dependent term in the activation energy R (4), (9) is linear in h, the problem (12) reduces to a linear equation for h. We give the solution of this equation for optimal control of the nucleation rate by a spatially uniform pulsed field $h(t)$ with a pulse duration τ, and the penalty functional is $G[h] = (1/2)\int_0^\tau dt\, h^2(t)$:

$$R_{\rm opt} \approx R^{(0)} - \left[2\mathcal{G}\kappa(\tau)\right]^{1/2}, \; \kappa(\tau) = \max_{t_c} \int_0^\tau dt\, \chi^2(t - t_c),$$
$$\chi(t) = \int d\mathbf{x}\, \chi_{\rm nucl}(\mathbf{x}, t), \qquad (13)$$
$$h_{\rm opt}(t) = -\chi(t - t_{cm})\left[2\mathcal{G}/\kappa(\tau)\right]^{1/2} \;\text{ for } 0 < t < \tau,$$

where t_{cm} is the value of t_c which provides the maximum to the function $\kappa(\tau)$ (13).

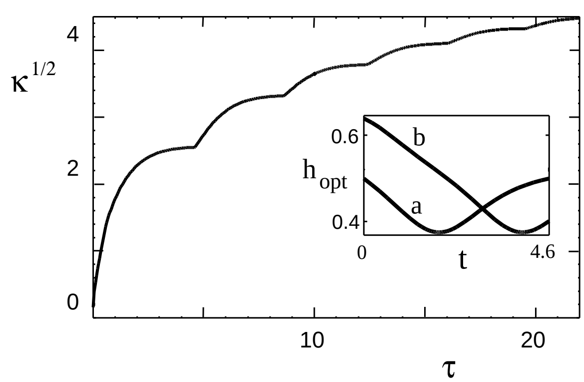

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FIG. 2. Reduced correction $\kappa^{1/2}(\tau)$ (13) for optimal control of the escape rate of a Brownian particle in a cosine potential, $\ddot{q} + 2\Gamma\dot{q} - \sin q = \xi(t)$, for $\Gamma = 0.04$. Inset: the optimal fields (13) for the pulse duration τ just below (a) and above (b) the switching value $\tau \approx 4.6$.

The above results can be easily reformulated for systems other than those described by the model (1). In particular, they apply to lumped parameter fluctuating systems, including over- and underdamped Brownian particles. For such systems, instead of the susceptibility for nucleation one should consider the susceptibility for *escape from a metastable state.*

In optimal control of escape of an underdamped Brownian particle by a pulsed field, the *shape* of the control field may change discontinuously with the varying pulse duration. This is related to the fact that the integral in the expression for $\kappa(\tau)$ may have several extrema for different t_c, and with the varying τ there may occur switchings between different extrema (cf. Fig. 1). Respectively, the activation energy is nondifferentiable at such τ, as shown in Fig. 2.

The above approach, including the formulation of the optimal control problem in terms of logarithmic susceptibility, applies also to systems away from thermal equilibrium if the driving noise is Gaussian. The logarithmic susceptibility with respect to the field h can still be expressed in terms of the optimal paths for $h = 0$, although the appropriate expressions differ in form from Eqs. (5), (9).

In conclusion, we have provided the nonadiabatic theory of escape and nucleation rates in systems driven by time-dependent fields. The effect of the field on the probabilities of large fluctuations has been described in terms of the logarithmic susceptibility. This susceptibility can be measured experimentally even if the underlying dynamics of the system is not known. It has been used to formulate the problem of optimal control. We have demonstrated singular behavior of optimal modulation of the escape rate as a function of the parameters of the control field.

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